

## Session VI

# ATOMIC MOTIONS AND DEBYE-WALLER FACTORS

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## On the Theory of Resonant Scattering of Gamma Rays by Nuclei Bound in Crystals\*

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### I. INTRODUCTION

An attempt is made in the present paper to give a physical description of the process in which a gamma quantum is resonantly scattered or absorbed by nuclei bound in a crystal structure. The discussion includes a description of the time development of these processes as well as a critical study of the limits imposed upon such a description by the laws of quantum mechanics.<sup>1</sup>

### II. ROLE OF THE CONSERVATION LAWS OF MOMENTUM AND ENERGY

The processes of emission, absorption, and scattering of radiation by nuclei bound in crystals are governed by the conservation laws of energy and momentum.

\* Presented by R. L. Mössbauer.

<sup>1</sup> For recent compilation of the literature relevant to this paper the reader is referred to review papers by A. J. F. Boyle and H. E. Hall [Rept. Progr. Phys. 25, 441 (1962)] and by R. L. [Mössbauer, Ann. Rev. Nucl. Sci. 12, 123 (1962)].

We first consider relations which exist between the momentum transferred to a nucleus during the emission of a gamma quantum and the corresponding energy transfer.

#### A. Free Recoiling Nucleus

The difference in the kinetic energy of the nucleus before and after the gamma emission  $\Delta E$  is given in both the classical and quantum-mechanical description of the process by an expression of the form

$$\begin{aligned}\Delta E &= (\mathbf{p} - \hbar\mathbf{k})^2/2m - \mathbf{p}^2/2m \\ &= (\hbar\mathbf{k})^2/2m - \mathbf{p} \cdot (\hbar\mathbf{k})/m,\end{aligned}\quad (1)$$

where  $|\mathbf{p}|$  is the momentum of the nucleus prior to the emission of the gamma quantum,  $\hbar\mathbf{k}$  is the momentum of the gamma quantum, and  $m$  is the mass of the nucleus. The first term in Eq. (1) is the recoil energy which would be given to a nucleus initially at rest, while the second term represents the Doppler

shift in the energy arising from the motion of the emitting nucleus.

### B. Nucleus Bound in a Crystal

In discussing this case from the *classical* point of view, we suppose for simplicity that the momentum  $\hbar\mathbf{k}$  is transferred to a single nucleus in a time short compared to the periods of the lattice vibrations. Under these circumstances the momentum is initially taken up by a single nucleus and then distributed throughout the whole crystal with the velocity of sound, via the binding forces. The center of mass of the crystal responds immediately to the momentum transfer. This is simply a consequence of the definition of the center of mass and therefore does not yield any physical description of the mechanism of the propagation of the disturbance through the crystal.

It is convenient to separate the total energy transferred to the crystal into a translational energy associated with the center of mass motion of the crystal and an internal energy depending on the relative positions and velocities of the constituent nuclei. The change  $\Delta E_T$  in the translation energy of the crystal arising from the emission of a gamma quantum of momentum  $\hbar\mathbf{k}$  is given by

$$\Delta E_T = \frac{(\mathbf{p}_M - \hbar\mathbf{k})^2}{2M} - \frac{\mathbf{p}_M^2}{2M} = \frac{(\hbar\mathbf{k})^2}{2M} - \frac{\mathbf{p}_M \cdot (\hbar\mathbf{k})}{M}, \quad (2)$$

where  $\mathbf{p}_M = \sum_j m_j \mathbf{v}_j$  is the center of mass momentum of the crystal,  $\mathbf{v}_j$  the velocity of the  $j$ th nucleus in the laboratory system, and  $M = \sum_j m_j$  is the total mass of the crystal. The total energy given to the crystal under the simplifying conditions introduced above is given by

$$\Delta E = (\hbar\mathbf{k})^2/2m_l - \mathbf{p}_l \cdot (\hbar\mathbf{k})/m_l, \quad (3)$$

where  $\mathbf{p}_l$  is the initial momentum of the emitting nucleus and  $m_l$  is its mass.

Thus the change in the internal energy  $\Delta E_I$  is given by

$$\Delta E_I = \Delta E - \Delta E_T = [(\hbar\mathbf{k})^2/2m_l - (\hbar\mathbf{k})^2/2M] - [(\mathbf{p}_l \cdot (\hbar\mathbf{k})/m_l - \mathbf{p}_M \cdot (\hbar\mathbf{k})/M)]. \quad (4)$$

The different terms represent, respectively, the recoil energy and the Doppler energy of the  $l$ th nucleus and of the center of mass of the crystal. The total energy transfer depends, according to Eq. (3), upon the momentum  $p_l$  possessed by the emitting nucleus at the time of emission of the gamma quantum. In the case of the freely recoiling nucleus this momentum could be determined, thus providing a well-defined relationship between the momentum and en-

ergy transfer to the nucleus. In the case of a bound nucleus, classical mechanics presents no barrier, at least in principle, to a determination of the initial momentum of the emitting nucleus at the time of the gamma emission. In sharp contrast to this classical conclusion, quantum mechanics does not permit a determination of the momentum of the emitting nucleus at the instant of emission. This is an immediate consequence of the fact that the nucleus, being bound in the crystal, is limited in its movement to a region in space of the order of interatomic distances. As a result the momentum has an uncertainty  $\Delta p_l \sim \hbar/a \sim 10^{-10}$  g cm/sec. On the other hand, in order to be able to distinguish between recoilless and nonrecoilless processes, we require a knowledge in the initial momentum of the emitting nucleus  $\Delta p_l \lesssim \Gamma m_l / |\hbar k| \sim 10^{-24}$  g cm/sec for a natural linewidth  $\Gamma$  of the nuclear excited state which is typically of the order of  $10^{-10}$  eV. These two conditions on  $\Delta p_l$  are clearly incompatible. The uncertainty in the initial momentum  $p_l$  of the emitting nucleus leads to an uncertainty in the energy transferred to the lattice which is so large that it becomes impossible to predict a *single emission process* for recoilless or nonrecoilless transitions. Quantum mechanics only permits the determination of *probabilities* for processes in which specific amounts of energy are transferred to the lattice. This is reflected by the fact that the final state wave function  $\Psi$  of the crystal is no longer an eigenfunction of the crystal Hamiltonian, but rather a linear superposition of such crystal eigenfunctions.

The relation between the crystal wave functions  $\psi_i$  and  $\Psi$  which describe the crystal before and after the gamma emission is given by

$$\Psi = e^{-i\mathbf{k} \cdot \mathbf{r}_l} \psi_i = \sum_n c_n \psi_n, \quad (5)$$

where the  $\psi_n$  are eigenfunctions of the crystal Hamiltonian  $H_C$ :

$$H_C \psi_n = E_n \psi_n. \quad (6)$$

The operator  $e^{-i\mathbf{k} \cdot \mathbf{r}_l}$  describes the transfer of momentum  $-\hbar\mathbf{k}$  to the crystal,  $\mathbf{r}_l$  being the position coordinate of the nucleus undergoing the transition. It should be emphasized that the coefficients  $c_n$  occurring in the expansion in Eq. (5) are independent of time as long as we limit attention to times long compared to the decay time of the nucleus undergoing the gamma transitions. Such conditions are imposed in the derivation of the Debye-Waller factor using time-dependent perturbation theory, as discussed in Sec. III.

Conservation of momentum in the gamma-emis-

sion process may be demonstrated by calculating the expectation values of the linear momentum operator for times long before and after the gamma emission. We show now that the momentum of the system consisting of a crystal + photon is conserved independently of the details of the actual momentum transfer and any associated transfer times.

If we suppose the crystal to be initially at rest we obtain for the expectation value of the linear momentum

$$(\psi, \mathbf{p}_C \psi) = (\phi, \varphi_i, \mathbf{p}_M \phi_i \varphi_i) + (\phi, \varphi_i, \sum \mathbf{p}_j \phi_i \varphi_i) = 0. \quad (7)$$

In writing Eq. (7) we have expressed the total wave function  $\psi$  of the crystal as a product of two factors, one depending on the center of mass coordinates  $\mathbf{r}_M$  of the entire crystal, the other on relative position coordinates  $\varphi_i$  of the individual nuclei. We have also used the fact that the operator representing the total internal momentum vanishes,  $\sum \mathbf{p}_j = 0$ ; hence we have for the total momentum operator of the crystal  $\mathbf{p}_C = \mathbf{p}_M + \sum \mathbf{p}_j = \mathbf{p}_M$ .

After the emission of a gamma quantum of momentum  $\hbar \mathbf{k}$ , we have

$$(\Psi, \mathbf{p}_C \Psi) = (e^{-i\mathbf{k} \cdot \mathbf{r}_i} \phi_i \varphi_i, \mathbf{p}_M e^{-i\mathbf{k} \cdot \mathbf{r}_i} \phi_i \varphi_i). \quad (8)$$

The position coordinate  $\mathbf{r}_i$  is that of the nucleus emitting the photon,  $\mathbf{r}_i = \mathbf{r}_M + \varrho_i$ . We then obtain

$$(\Psi, \mathbf{p}_C \Psi) = (\Psi, \mathbf{p}_M \Psi) = -\hbar \mathbf{k}. \quad (9)$$

Thus a momentum  $-\hbar \mathbf{k}$  equal in magnitude but with sign opposite to that of the gamma quantum is associated with the center of mass motion of the crystal. This result is intimately connected with the usual statement that the momentum is taken up by the whole crystal. This statement is in complete accord with the limitations on measurement imposed by quantum mechanics. The gamma emission causes a disturbance to propagate with the velocity of sound through the crystal. The spread of this disturbance can be discussed in terms of the propagation of sound waves through the crystal in the classical case or in terms of the propagation of phonons in the quantum-mechanical case.

In quantum mechanics, however, it is not possible to give a detailed account of the process by which momentum is distributed throughout the crystal, if at the same time we require a knowledge of *how much* energy and momentum has been transferred to the crystal. It can be shown that the uncertainty principle does not permit an experimental check as to whether a certain part of the crystal has taken up

the recoil momentum after a time characteristic for the propagation of sound throughout a given region of the crystal. In order to demonstrate this, let us first assume that only a limited part of the crystal takes up the recoil momentum. Classically, we would suppose that in a characteristic time  $\tau \sim 10^{-7}$  sec within which a typical gamma emission takes place, a disturbance could travel a distance  $L \sim \tau v_s \sim 10^{-2}$  cm, where  $v_s$  is the velocity of sound in the solid,  $v_s \sim 10^5$  cm/sec.  $L$  is the extension of the part of the crystal that classically has taken up the momentum. However, we see that in quantum mechanics the recoil energy of this part of the crystal cannot be determined, since this energy is considerably smaller than the zero-point energy which is associated with this portion of the crystal. This can be seen as follows.<sup>2</sup> Suppose that a portion of the crystal having mass  $\mu = (L/a)^3 m$  oscillates as a whole with a frequency  $\omega \sim \omega_L(a/L)$ , where  $\omega_L$  is a typical lattice vibration frequency,  $a$  is the interatomic distance, and  $m$  is the mass of a single nucleus. The square of its zero-point momentum is then given by

$$|p_\mu|^2 \sim 2\mu(\hbar\omega/4) \sim (1/2)(L/a)^2 m \hbar \omega_L. \quad (10)$$

One should bear in mind, that although the zero-point energy itself is precisely determined by the frequency  $\omega_L$ , one has to attribute fluctuations to the kinetic and the potential energy parts of the total zero-point energy which are of the order of the total zero-point energy itself.

In order to obtain a sizeable probability for recoilless transitions, the transferred momentum must fulfill the condition  $(\hbar \mathbf{k})^2 \lesssim m \hbar \omega_L$ . Since  $L \gg a$ , we have a resulting uncertainty in the kinetic energy of the portion of the crystal under consideration, which is of order  $|p_\mu|^2/m \gg (\hbar k)^2/2m$ , and it is not possible to verify by measurement that the recoil energy or momentum is actually carried by this portion of the crystal.

A time of at least  $\Delta t \sim (\hbar k^2/2M)^{-1}$  is required to measure the recoil energy of the *whole* crystal (which of course is not bound and therefore is not subject to zero-point fluctuations). This time, expressed in terms of the linear extension  $X$  of the whole crystal, is given by  $\Delta t \sim 2(X/a)^3(m/\hbar k^2) \sim 2(X/a)^3(1/\omega_L)$ . On the other hand, we have for the time in which the sound reaches the edge of the crystal:  $\Delta t_s = X/v_s$  or  $\Delta t_s \sim X/(a\omega_L/2\pi)$ . Consequently,

$$\Delta t/\Delta t_s \sim 4\pi(X/a)^2 \gg 1. \quad (11)$$

<sup>2</sup> V. F. Weisskopf, "The Mössbauer Effect," in *Lectures in Theoretical Physics* (Interscience Publishers, Inc., New York, 1961), Vol. III.

Thus, to measure the recoil energy of the crystal and to verify the conservation of energy will take a time long compared to the time necessary for the sound to propagate through the crystal. Therefore, the mechanism by which the momentum is distributed throughout the crystal is not accessible to observation and consequently the conclusion that the whole crystal takes up the momentum is not in conflict with the notion of a finite wave velocity of disturbances propagating through a crystal lattice. These remarks naturally apply only to the case where the zero-point energies associated with the vibrations of individual nuclei inside the crystal exceed or are comparable to the average recoil energy given to the crystal, the situation prevailing in the cases of interest here. If one drops this restriction and deals instead with cases where the average recoil energies are much larger than the zero-point energies of the individual nuclei, one can of course follow in detail experimentally the spread of the disturbance throughout the crystal.

The preceding discussion of the process of energy and momentum transfer to the lattice can be summarized as follows: The momentum is taken up by the whole crystal. The energy associated with the translational motion of the center of mass is negligible. The internal energy transitions, on the other hand, occur as phonon transitions between lattice oscillator states which, however, do not store momentum as the expectation value of the momentum operator for any oscillator state is zero. We therefore have the situation that the momentum is picked up by the whole crystal, without any significant amount of accompanying energy going to the center of mass, while the energy is picked up by the lattice oscillators, without any corresponding momentum transfer to the oscillators. It is in this sense that we may consider momentum and energy as decoupled quantities.

### III. RECOILLESS RESONANCE ABSORPTION

#### A. Qualitative Considerations

The general conditions prerequisite for a large probability of recoilless emission or absorption are easily understood from the following qualitative considerations: An atom bound in a solid is confined to a region in space of linear extension  $\Delta x$  smaller than the interatomic distances. This leads to a corresponding uncertainty in momentum  $\hbar|\Delta\mathbf{k}| \gtrsim \hbar/\Delta x$ . In case the momentum  $\hbar\mathbf{k}$  transferred during a gamma transition to the crystal is comparable with or smaller than  $\hbar\Delta\mathbf{k}$ , it is no longer possible to predict recoilless

or nonrecoilless transitions in a single process. The inequality  $|\mathbf{k}| \lesssim 1/\Delta x$  thus constitutes the condition prerequisite for the existence of a large probability for recoilless transitions. This condition can easily be formulated in a somewhat more quantitative way if we make use of the fact that the recoil energy given to a nucleus bound in a crystal equals in the *average* the recoil energy received by a free recoiling nucleus in a single process. This result follows from both a classical and a quantum-mechanical consideration. In the classical case this may be seen, for instance, from our Eq. (3), where the Doppler term vanishes if we average over a large ensemble of particles leaving as the average energy transferred the recoil energy of a single nucleus, initially at rest and free to recoil. Thus we have  $\langle\Delta E\rangle_{av} = (\hbar\mathbf{k})^2/2m$ . Let us now consider a very simple model in which we describe the internal motions in the crystal by a single frequency  $\omega_E$ , the so-called Einstein frequency. It is instructive to consider the two limiting cases in which the average recoil energy is either large or small compared to the transition energy of the Einstein oscillators:

- (i)  $\langle\Delta E\rangle_{av} \gg \hbar\omega_E$
- (ii)  $\langle\Delta E\rangle_{av} \ll \hbar\omega_E$ .

In case (i), many transitions of Einstein oscillators are on the average required to transfer an energy contribution of order  $\langle\Delta E\rangle_{av}$  to the lattice. Consequently, the probability of a nuclear transition taking place without simultaneous phonon transitions will be small. The opposite situation prevails in case (ii), where it is evident that nuclear transitions will only rarely be associated with simultaneous phonon transitions, which results in a high probability for recoilless transitions. The situation is illustrated in Fig. 1. These qualitative considerations apply equally well to a real crystal, if one replaces the Einstein frequency  $\omega_E$  by a characteristic frequency of the vibration spectrum of the crystal, typically of the order of  $\kappa\theta/\hbar$ , where  $\theta$  is the Debye temperature. We then obtain as the essential condition which leads to a high probability for recoilless gamma transitions  $(\hbar\mathbf{k})^2/2m_L \lesssim \kappa\theta$ .

#### B. Debye-Waller Factor

The Debye-Waller factor is defined as the fraction of transitions occurring as recoilless gamma transitions of nuclei which are bound in a crystal and are initially in thermal equilibrium. It should be emphasized that the term "recoilless" specifies only the absence of energy transfers to the lattice; momentum, of course, is always transferred in both the re-

coilless and the nonrecoilless transitions. Certain features of the Debye–Waller factor can easily be understood on the basis of simple semiclassical arguments: An electromagnetic wave train emitted or scattered by a nucleus suffers a frequency modulation as a result of the vibrational motion of the nucleus around its equilibrium position. This frequency modulation leads to a central unshifted line and to side components. The central line (frequency  $\omega_0$ ) represents the recoilless processes and one can define the Debye–Waller factor as the fractional in-

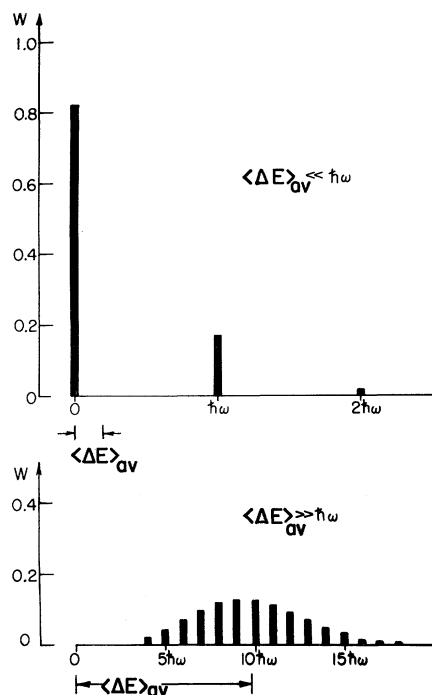


FIG. 1. Relative probability  $W$  for a gamma transition accompanied by simultaneous (one phonon) transitions of 0, 1, 2, . . .  $n$  Einstein oscillators of frequency  $\omega$ , at the temperature  $T = 0$ , illustrated for two different values of the average energy transfer  $\langle \Delta E \rangle_{av}$ . [From R. L. Mössbauer, Les Prix Nobel EN 1961, Nobel Foundation, Stockholm, 1962; translation: Science **137**, 731 (1962).]

tensity of the central line. The side components represent those gamma transitions which are accompanied by phonon transitions. Care must be exercised, however, in carrying this classical description too far. The description of the central line is possible in a semiclassical way, if one incorporates the zero-point motions of the nuclei into the picture.<sup>3</sup> On the other hand, the description of the side components on the basis of semiclassical arguments fails entirely.

<sup>3</sup> F. L. Shapiro, Usp. Fiz. Nauk **72**, 685 (1960) [English transl.: Soviet Phys.—Usp. **4**, 881 (1961)].

It becomes obvious that quantum effects play a major role in the description of the side components if one considers that frequencies  $\omega_0 \pm \Delta\omega$  appear with equal intensity in the classical frequency modulation picture, thus giving equal probabilities for transitions with frequency changes  $\pm \Delta\omega$ , whereas such a situation prevails in quantum mechanics only in the limit of high phonon occupation numbers. The classical description fails in the presence of low phonon occupation numbers, the situation prevailing in the cases of interest here.

The transition amplitude for the scattering of a gamma quantum with initial momentum  $\hbar\mathbf{k}_i$  into a state with final momentum  $\hbar\mathbf{k}_f$ , while the lattice undergoes a transition from the initial state  $|i\rangle$  through an intermediate state  $|m\rangle$  to a final state  $|f\rangle$  which is assumed to be identical with the initial state  $|i\rangle$ , is according to Lamb<sup>4</sup> proportional to

$$f_{fi} = \sum_m \frac{\langle i | e^{-i\mathbf{k}_f \cdot \mathbf{u}} | m \rangle \langle m | e^{i\mathbf{k}_i \cdot \mathbf{u}} | i \rangle}{E_i - E - (\epsilon_m - \epsilon_i) + i\Gamma/2}. \quad (12)$$

In this equation  $E_i$  denotes the ground-state energy of the nucleus undergoing the transition,  $\epsilon$  denotes lattice energies,  $E - i\Gamma/2$  denotes the “resonance” energy,  $\Gamma$  is the total width of the intermediate state (which in good approximation can be assumed to be independent of the state of the lattice), and  $\mathbf{u}$  is the position coordinate relative to its equilibrium position of the particular nucleus undergoing the transition. The operators  $e^{\pm i\mathbf{k} \cdot \mathbf{u}}$  appearing in Eq. (12) result from the expansion in terms of plane waves of the vector potential  $\mathbf{A}$  in the operator  $[\mathbf{p} - (e/c)\mathbf{A}]^2$  which describes the interaction of the nucleus bound in the lattice with the electromagnetic field. Equation (12) contains matrix elements which depend on the position coordinates of the center of masses of the constituent nuclei in the lattice. The nuclear matrix element, which depends only on the internal coordinates of the nucleons inside the nucleus and which should appear as a factor of the lattice matrix elements, was dropped in Eq. (12). The factorization of the nuclear and lattice matrix elements becomes possible as a result of (1) the mutual independence of the lattice states upon the internal nuclear coordinates and the nuclear states upon the lattice coordinates and (2) of the fact that typical transition energies in the lattice (phonons) are small in comparison with nuclear transition energies.

We consider Eq. (12) in the two extreme cases in which the total width  $\Gamma$  of the intermediate level

<sup>4</sup> W. E. Lamb, Phys. Rev. **55**, 190 (1939).

(resonance level) is either large or small compared to typical energy transfers to the lattice:

$$(i) \quad |E_i - E + i\Gamma/2| \gg |\epsilon_m - \epsilon_i| \quad (13)$$

$$(ii) \quad |E_i - E + i\Gamma/2| \ll |\epsilon_m - \epsilon_i|. \quad (14)$$

We obtain in these two cases the results:

$$(i) \quad f_{ii} = \frac{\langle i | e^{-i(\mathbf{k}_f - \mathbf{k}_i)\mathbf{u}} | i \rangle}{E_i - E + i\Gamma/2} \quad \text{fast collision} \quad \text{(x-ray scattering)} \quad (15)$$

$$(ii) \quad f_{ii} = \frac{\langle i | e^{-i\mathbf{k}_f\mathbf{u}} | i \rangle \langle i | e^{i\mathbf{k}_i\mathbf{u}} | i \rangle}{E_i - E + i\Gamma/2} \quad \text{slow collision} \quad \text{(gamma-resonance scattering)}. \quad (16)$$

The justification for the classification of the two cases given by Eqs. (15) and (16) as "fast" and "slow" collisions follows from an investigation of the time dependence of the displacement operator  $\mathbf{u}$  during the scattering process.<sup>5</sup> Following Van Hove<sup>6</sup> and Singwi and Sjölander<sup>7</sup> we rewrite Eq. (12)<sup>8</sup>:

$$\begin{aligned} f_{ii} &= \sum_m \langle i | e^{-i\mathbf{k}_f\mathbf{u}} | m \rangle \langle m | e^{i\mathbf{k}_i\mathbf{u}} | i \rangle \\ &\quad \times \int_{-\infty}^{+\infty} \frac{\delta[\rho - (\epsilon_m - \epsilon_i)]}{(E_i - E - \rho) + i\Gamma/2} d\rho \\ &= (1/2\pi) \sum_m \int_0^\infty dt \langle i | e^{-i\mathbf{k}_f\mathbf{u}} | m \rangle \langle m | e^{i\mathbf{k}_i\mathbf{u}} | i \rangle e^{i(\epsilon_i - \epsilon_m)t} \\ &\quad \times \int_{-\infty}^{+\infty} \frac{e^{i\rho t}}{(E_i - E - \rho) + i\Gamma/2} d\rho \\ &= (1/i) \int_0^\infty dt e^{+it(E_i - E) - \Gamma t/2} \sum_m \langle i | e^{-i\mathbf{k}_f\mathbf{u}} e^{-iH_C t} | m \rangle \\ &\quad \times \langle m | e^{i\mathbf{k}_i\mathbf{u}} e^{iH_C t} | i \rangle \\ &= (1/i) \int_0^\infty dt e^{+it(E_i - E) - \Gamma t/2} \langle i | e^{-i\mathbf{k}_f\mathbf{u}(t)} e^{+i\mathbf{k}_i\mathbf{u}(0)} | i \rangle, \quad (17) \end{aligned}$$

where  $u(t)$  is the displacement operator in the Heisenberg representation. The time  $t$  appearing in the operators in Eq. (17) can be interpreted as a "collision time" within which the momenta  $\mathbf{k}_i$  and  $-\mathbf{k}_f$  are transferred.<sup>9</sup> This collision time is on the average of the order of the lifetimes of the relevant intermediate states. On the basis of this interpretation it is possible to evaluate the integral in Eq. (17) for special cases, where the matrix element in the integral becomes independent of the time  $t$ .<sup>9</sup> The correlation between the displacement operators  $\mathbf{u}(t)$  and  $\mathbf{u}(0)$  becomes

particularly simple in these special cases. Of particular significance are the following two cases:

(1) For lifetimes  $\tau$  of the intermediate states which are short in comparison with lattice vibration times, the position coordinates do not change during the scattering process and one may set  $\mathbf{u}(t) \approx \mathbf{u}(0)$ . Equation (17) then reduces to Eq. (15), which thus represents the case of "fast" scattering. Such a situation exists in the case of the scattering of x rays from atoms bound in a crystal. The intermediate states in this case are states in the optical continuum with lifetimes  $\tau$  very short compared to typical lattice vibration times  $\tau_L$  ( $\tau_L \sim 10^{-13}$  sec).

(2) In the opposite case of a lifetime  $\tau$  of the intermediate state which is long in comparison with typical lattice vibration times, the correlation between position coordinates vanishes entirely and we may set  $u(t) \approx u(\infty)$ . We use the relation<sup>6</sup>  $\{\exp(-i\mathbf{k}_f\mathbf{u}) \exp(i\mathbf{k}_i\mathbf{u})\}_T = \{\exp(-i\mathbf{k}_f\mathbf{u})\}_T \times \{\exp(i\mathbf{k}_i\mathbf{u})\}_T$ , which applies to a crystal in thermal equilibrium, i.e., to a situation where the initial states are thermally averaged. Equation (17) then reduces to the expression given by Eq. (16), which thus represents the case of "slow" scattering. This is the situation prevailing in the case of recoilless gamma resonance scattering, where the lifetime  $\tau$  of the intermediate states (nuclear excited levels) is always long in comparison with lattice vibration times.

It should be observed in this context that in arriving at Eq. (16) we have used the thermal average of the scattering amplitude rather than the thermal average of its square, a procedure which applies only in the case where the incoherent elastic scattering can be neglected compared to the coherent elastic scattering.<sup>5</sup>

The transition probabilities (Debye-Waller factors) for the two cases considered above are given directly by the squares of the matrix elements appearing in Eq. (17), with a thermal average performed over the initial states. One then obtains for x-ray scattering and gamma resonance scattering, respectively,

$$e^{-2W} = \{ |\langle i | e^{-i(\mathbf{k}_f - \mathbf{k}_i)\mathbf{u}} | i \rangle|^2 \}_T \quad \text{x-ray scattering} \quad (18)$$

$$e^{-2W_f} e^{-2W_i} = \{ |\langle i | e^{-i\mathbf{k}_f\mathbf{u}} | i \rangle|^2 \}_T \{ |\langle i | e^{i\mathbf{k}_i\mathbf{u}} | i \rangle|^2 \}_T \quad \text{gamma resonance scattering}. \quad (19)$$

Equation (19) involves a product of two Debye-Waller factors, since it was obtained under the assumption that both the absorbed and re-emitted quanta are involved in recoilless transitions. Under usual experimental conditions one does not measure

<sup>5</sup> G. T. Trammel, Phys. Rev. 126, 1045 (1962).

<sup>6</sup> L. Van Hove, Phys. Rev. 95, 249 and 1374 (1954).

<sup>7</sup> K. S. Singwi and A. Sjölander, Phys. Rev. 120, 1093 (1960).

<sup>8</sup> The same transformation has already been used by Lamb (Ref. 4) without, however, introducing the notion of time.

<sup>9</sup> G. C. Wick, Phys. Rev. 94, 1228 (1954).

the final state, thus limiting only the incident quantum to recoilless transitions. Under such conditions the product of Debye-Waller factors appearing in Eq. (19) reduces to the single factor  $e^{-2W_i}$ .

#### IV. ON MOMENTUM TRANSFER TIMES

It has been asserted<sup>10,11</sup> that when a gamma quantum is scattered by a nucleus bound in a crystal, the momentum can be considered as being transferred in two "sudden" pieces, one corresponding to the absorption of an incoming photon, the other to the emission of an outgoing photon. This description is sometimes suggested<sup>11</sup> on the basis of the product form of the Debye-Waller factors occurring in Eq. (19). We feel, however, that this description is not completely in accord with the quantum mechanics as usually interpreted and furthermore that it is not necessary for a clear understanding of the physics of nuclear resonance scattering of gamma radiation. Our position rests on the following considerations:

(a) The recoilless *scattering* of gamma rays is not sudden in the sense of the impulse approximation.<sup>12,13</sup> A fundamental requirement for the applicability of the impulse approximation is that the lifetime  $\tau$  of the intermediate state should be short compared to the lattice periods,  $\tau \ll 1/\omega_L$ . This condition is surely not fulfilled in the case of interest here ( $\omega_L \sim 10^{13} \text{ sec}^{-1}$ ,  $\tau \sim 10^{-7} \text{ sec}$ ).<sup>14,15</sup> In fact, we have to do here with a case of "strong binding" in the sense of Lamb.<sup>4</sup> The opposite case of "weak binding" is illustrated by the scattering of photons by a proton moving freely in a cloud chamber. The track of the proton shows a rather abrupt change in its direction at the position where the scattering event occurred. This phenomenon indicates a sudden transfer of the momentum. Here, in contrast to recoilless resonance scattering by bound nuclei, we have a true weak binding process, the level spacing of a free proton being approximately zero. The impulse approximation therefore applies. These considerations also reflect the circumstance that the specific physical properties of the system under consideration always enter

in a crucial way into any discussion of the measurability of the observables of the system.

(b) It is true that when a gamma quantum is absorbed or emitted energy and momentum are transferred to and from the nucleus in indivisible units. In this respect gamma quanta have particle-like properties. It is most tempting to say that since one piece only of momentum is transferred, it must be transferred suddenly. But to ask about the time development of transition processes is to ask for a knowledge of quantities complementary to energy and momentum—and it must be realized that quantum mechanics imposes limitations on the knowledge of these quantities, a knowledge which has to be obtained by observation.

Consider a wave packet of electromagnetic radiation representing just one quantum, which passes over a group of nuclei. Such a pulse contains a distribution of frequencies  $\Delta\omega$ . The probability for the absorption of a gamma quantum by a nucleus is proportional to the square of the field amplitude and is significantly large only while the wave packet has a large amplitude in the space around the nuclei. The associated time interval is  $\Delta t \sim 1/\Delta\omega$ . Now, there is no way of saying at what instant within this time interval the gamma quantum is transferred from the radiation field to the nucleus or atom that absorbs it.  $\Delta t$  may be large or small depending on how the radiation pulse was prepared.

One might argue that in nuclear resonance absorption one only requires the knowledge of energies to within the order of nuclear transition energies  $E_\gamma$ , typically  $10^5 \text{ eV}$ . One can prepare a state, for example by delayed coincidence selection, for which  $\Delta t \sim \hbar/E_\gamma \sim 10^{-20} \text{ sec}$  which is indeed instantaneous compared to any other times in the problem. But this is a consequence of the special initial state which was prepared and does not support the idea that absorption and emission processes are *in general* happening with times of the order of  $10^{-20} \text{ sec}$ . In particular, no such conditions hold for the *average* incident wavepacket which has a width  $\Delta t \sim \hbar/\Gamma$ . This appears even more clearly when we turn from absorption to emission. Here the probability (for emission) is usually proportional not to an expression whose time dependence is sharply peaked (incident pulse of gamma radiation) but to a decaying exponential  $\exp(-\Gamma t/\hbar)$ . According to time-dependent perturbation theory this amplitude describes the probability that a transition from an appropriately prepared<sup>16</sup> initial state  $|i\rangle$  to a final state  $|f\rangle$  has occurred in some

<sup>10</sup> J. Petzold, Sitzber. Heidelberg. Akad. Wiss., Math. Naturw. Kl., Abhandl. 5 (1961); Z. Physik 163, 71 (1961); *Proceedings of the Second Mössbauer Conference, Paris* (John Wiley & Sons, Inc., New York, 1962), p. 2.

<sup>11</sup> H. J. Lipkin, Ann. Phys. 18, 182 and 294 (1962).

<sup>12</sup> G. F. Chew, Phys. Rev. 80, 196 (1950).

<sup>13</sup> G. F. Chew and G. C. Wick, Phys. Rev. 85, 636 (1952).

<sup>14</sup> A somewhat less stringent condition for the validity of the impulse approximation has recently been proposed by Goldberger and Watson (Ref. 15). This condition is likewise not fulfilled in the case of low-energy gamma resonance scattering.

<sup>15</sup> M. L. Goldberger and K. M. Watson, *Collision Theory* (John Wiley & Sons, Inc., New York, to be published), Chap. 11.

<sup>16</sup> M. Gell-Mann and M. L. Goldberger, Phys. Rev. 91, 398 (1953).

finite time interval  $t > 0$ . This amplitude determines the probability to find the system in a certain final state at a certain time—but the details of how or when the system reached that state are not ordinarily accessible to observation.

Of course, one can measure the emitted gamma ray and thus determine that the decay has occurred in a certain interval of time. Furthermore, one can select experiments which detect only those gamma rays emitted in times much shorter than the half-life  $\tau$  of the nuclear excited state involved ( $\tau \sim 10^{-7}$  sec). In resonance scattering such emission processes, however, constitute only a small fraction of those processes which are of experimental relevance. But in selecting such fast processes as those representing the time of momentum transfer in emission *in each single case*, the description of nature is being carried beyond the limits of quantum mechanics.

One should note that the arguments presented above, showing that one can not infer the suddenness of the momentum transfer in a general process from a knowledge of the shape of the incident radiation pulse, do not vitiate the conclusion reached in part (a) with regard to the cloud-chamber example. In that case the suddenness of the momentum transfer could be experimentally verified by observing the sharp bend in the track of the free proton, a type of measurement not available in the case of low energy resonance scattering by nuclei bound in crystals, where it is in general not even possible to determine which particular nucleus received the momentum.

(c) The evaluation of zero-phonon transition

probabilities for gamma resonance scattering involves an integration over times of the order of the lifetime of the nuclear state involved, as has been pointed out by Inglis<sup>17</sup> and by Boyle and Hall.<sup>18</sup> This shows that there is no need for a concept of a sudden momentum transfer and that there is sufficient time for the recoil momentum to be transferred to the lattice via the binding forces during the scattering process.

(d) The transfer of the recoil energy to a single nucleus or a few nuclei making up a piece of the crystal can not be measured on account of the zero point energies—as pointed out in Sec. II. Moreover, the times involved in verifying the conservation of energy and momentum by measuring these quantities are long compared to the time of propagation of disturbances through the crystal at the velocity of sound. It therefore is difficult to see how the impulse momentum transfer picture can, from the quantum-mechanical point of view, contribute to an understanding of how momentum is transferred to the crystal.

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<sup>17</sup> D. R. Inglis, in H. Frauenfelder, *The Mössbauer Effect* (W. A. Benjamin, Inc., New York, 1962), p. 169.

<sup>18</sup> A. J. Boyle and H. E. Hall, *Rept. Progr. Phys.* **25**, 441 (1962).

## Lattice Dynamical Aspects of the Resonance Absorption of Gamma Rays by Nuclei Bound in a Crystal

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It is quite common in studies of the resonant emission or absorption of gamma rays by nuclei bound in a crystal that the emitting (or absorbing) nucleus is either an impurity in a host crystal or the crystal is itself imperfect. When this is the case, it is necessary in analyzing the results of such experiments to take into account the modification of the lattice vibrations of the host crystal caused by its departure from ideality. These modifications can give rise to interesting effects in the absorption spectrum of the perfect host crystal and, once the origins of these effects

are understood, a proper analysis of experimental results can yield worthwhile information about the dynamics of the host crystal and of the impurity atom.

At the present time there appear to be three dynamical quantities of interest in connection with the resonant absorption of gamma rays by nuclei in crystals. These are the “no-phonon” absorption cross section, the “one-phonon” absorption cross section, and the second-order Doppler shift. The first of these is the cross section for processes in which the